OIPE IAP40
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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:	Ramji Srinivasan, et al.	Atty. Docket No.:	5242.00120
Confirmation No.	7275		
Application No.:	10/661,768	Group Art Unit:	1731
Filed:	September 15, 2003	Examiner:	Tatyana Zalukaeva
For:	FORMALDEHYDE FREE INSULATION BINDER		

DECLARATION UNDER 37 C.F.R. §1.131

U.S. Patent and Trademark Office
Customer Service Window
401 Dulany Street
Alexandria, VA 22314

Dear Sir:

We, Ramji Srinivasan, Cornel Hagiopol and Natasha R. Bailey, do hereby aver and state as follows:

1.) THAT we are named as co-inventors of the subject U.S. patent application;

2.) THAT all of the events and all of the related testing identified in this declaration occurred prior to August 16, 2002.

3.) THAT after we had earlier conceived of the invention, and as a consequence of the events and related testing identified in this declaration, we reduced our invention, as it is described and claimed in the above-captioned patent application, to practice in this country before August 16, 2002.

4.) THAT all of the Attachments referred to hereinafter are copies of original laboratory notebook records which have been retained as a regular part of and in the ordinary course of Georgia-Pacific Resins, Inc.'s, (hereinafter GPRI's) business. It is the

practice of GPRI to require its employees who are involved in conducting the kind of activities referred to hereinafter to maintain and retain laboratory notebooks as a record of such activities.

5.) THAT as evidenced by the laboratory notebook records of Attachment A, a water soluble copolymer of maleic anhydride (MA) and hydroxyethyl acrylate (HEA) was prepared by free radical polymerization in the presence of allyloxypropane diol (APD) as a chain transfer agent. As shown in Attachment A, 2.753 moles of maleic anhydride were added into a reaction vessel containing water. The reaction vessel was equipped with a charging funnel. The contents of the reactor were heated to 72 °C and held at that temperature for 30 minutes while a solution of 2.066 moles of hydroxyethyl acrylate in water was prepared. 45 parts (0.3404 moles) of the chain transfer agent allyloxypropane diol and 2.5 parts of the free radical catalyst azodiisobutyronitrile (AIBN) then were added to the reactor. Over a period of about 90 minutes, the hydroxyethyl acrylate solution was added into the reactor to conduct the polymerization. Once the HEA was added, the reaction was allowed to remain at the elevated temperature for an additional 30 minutes before cooling was conducted. The relative mole ratio MA:HEA was 1.33:1.0 to provide a mole ratio of –COOH:-OH of about 2.7:1.0. The resin product had a solids content of about 32 wt. %.

6.) THAT as evidenced by the laboratory notebook records of Attachment B, another water soluble copolymer of maleic anhydride (MA) and hydroxyethyl acrylate (HEA) was prepared by free radical polymerization in the presence of allyloxypropane diol (APD) as the chain transfer agent and AIBN as the free radical polymerization catalyst. The process of preparing the copolymer was similar to that described above in paragraph 4.).

7.) THAT, as evidenced by the laboratory notebook records of Attachment C, (notebook record 236G45 and notebook record 236G46) two additional water soluble copolymers of maleic anhydride (MA) and hydroxyethyl acrylate (HEA) were prepared by free radical polymerization, this time in the presence of mercapto ethanol (also known as thio glycol) as the chain transfer agent. In addition, a small amount of a cationic monomer, [2-(methacrloyloxy)ethyl] trimethylammonium chloride (MTA) was added to yield a terpolymer. The free radical processes of preparing the two terpolymers were similar to that described above in paragraph 4.) as described for the preparation of the MA-HEA copolymer, with the MTA being added to the reactor along with the HEA. A different amount of the chain transfer agent was used in each synthesis. For both syntheses, the relative mole ratio of the three monomers was about MA:HEA:MTA of 0.97:1.0:0.16 to provide a mole ratio of –COOH:-OH of about 1.9:1.0. In the first synthesis (first page of exhibit), we measured the solids content of the resin product to be about 29.6 wt. %. In the second synthesis, we measured the solids content of the resin product to be about 29.5 wt. %.

8.) THAT Attachment D is a laboratory notebook record (notebook page 228G50) of the free radical polymerization of maleic anhydride (MA) and hydroxyethyl acrylate (HEA) in the presence of sodium-1-allyloxy-2-hydroxypropyl sulfonate (COPS) as the chain transfer agent. As shown in Attachment D, 0.52 mole of maleic anhydride was added into a reaction vessel containing water. The contents of the reactor were heated to 72 °C and held at that temperature for 20 minutes. About 20 parts (0.05 mole) of the chain transfer agent, COPS then was added. 0.7 parts of the free radical catalyst azodiisobutyronitrile (AIBN) was added and then over a one

hour period, 0.78 mole of hydroxyethyl acrylate was added into the reactor. Once the addition of the HEA was complete, the reaction was allowed to remain at the elevated temperature for an additional 30 minutes. Cold water was added to stop the reaction yielding a resin product with a solids content of about 15 wt. %. The relative mole ratio of MA:HEA was 0.67:1.0 to provide a mole ratio of –COOH:-OH of about 1.3:1.0.

9.) THAT the water soluble resin prepared in Attachment D was used thereafter as a binder to prepare handsheets from glass fibers and the tensile (strength) properties of the handsheets were measured. Hand sheets were prepared by sprinkling a binder composition comprising the soluble resin of Attachment D onto a glass mat previously formed by dewatering ½ inch PPG M-8035 chopped glass fibers dispersed in water, containing a polyacrylamide dispersing agent, through a screen. Excess binder was vacuumed off the glass fibers and then the binder-treated sheet was cured in an oven at 220° C for a set period of time of from 1 to 5 minutes to cure the binder and produce a small glass fiber mat for testing.

10.) THAT both dry tensile and hot/wet tensile strengths were measured for the handsheets (mats) prepared using the binder of paragraph 8.) and other binders that are reported below. Dry tensile strengths of handsheet (mat) samples (3 inches by 5 inches) were measured using a QC-1000 Materials Tester by the Thwing Albert Instrument Co. The hot/wet tensile properties of such mates were measured by soaking the so-prepared handsheets in 185° F (85° C) water for 10 minutes. Samples of the handsheets (3 inches by 5 inches) were then subjected to breaking using the QC-1000 Materials Tester by the Thwing Albert Instrument Co. while the handsheets were still hot and wet.

11.) THAT Attachment E (notebook records 228G51, 228G52 and 228G53) shows the calculations for preparing several binder formulations using various resin products for preparing handsheets (identified by the notebook record of their synthesis). Attachment E also

provides information about the handsheets and the results of the testing referred to in paragraphs 9.) and 10.). For purpose of the present invention, the binder labeled 5 (Binder 5) was prepared using the resin prepared in accordance with paragraph 8.) (Resin 228G50). 500 grams of a binder composition containing 5 wt. % resin solids were prepared by mixing 166.33 g of the 15.03 wt. % water soluble resin of Attachment D with 333.67 g of water. The calculation for preparing the 5% binder is shown (and highlighted in yellow) in the lower right corner of the first page of Attachment E. The second and third pages of Attachment E present the tensile test results for "Binder 5" (far right columns) The raw data for the dry tensile testing of 14 replicates is setoff with yellow highlighting on page 2 and the hot/wet tensile testing of 14 replicates is setoff with yellow highlighting on page 3 and respectively correspond to average tensile strengths of 26.1 pounds and 3.7 pounds. Based on these results we concluded that the resin had been successfully used and thus was useful as a binder for glass fibers.

12.) THAT as evidenced by the laboratory notebook records of Attachment F (notebook record 228G64), a water soluble copolymer of itaconic acid (IA) and hydroxyethyl acrylate (HEA) was prepared by free radical polymerization in the presence of allyloxypropane diol as the chain transfer agent. As shown in Attachment F, about 0.14 mole of allyloxypropane diol and about 0.59 mole of itaconic acid were added into a reaction vessel containing water. The contents of the reactor were heated to dissolve the itaconic acid and then to 72 °C and held at that temperature for about 20 minutes. 0.7 parts of the free radical catalyst azodiisobutyronitrile (AIBN) was added and over a one hour period, about 0.80 mole of hydroxyethyl acrylate was added into the reactor. Once the HEA was added, the reaction was allowed to remain at the elevated temperature for an additional four hours, at which point the temperature was raised to 76 °C and then an additional 0.21 part of AIBN was added. Thereafter, an aqueous solution of a resin having a solids content of about 21 wt. % was recovered. The

relative mole ratio of the two monomers was IA:HEA of 0.74:1.0 to provide a mole ratio of –COOH:-OH of about 1.5:1.0.

13.) THAT Attachment G (notebook records 228G65 and 228G66) documents the preparation of several binder formulations for preparing handsheets and the results of conducting the testing referred to in paragraphs 9.) and 10.) on the resulting handsheets. For purpose of the present invention, the binder labeled 5 ("Binder 5") was prepared using the resin prepared in accordance with paragraph 12.). 400 grams of binder containing 15 wt. % resin solids was prepared by mixing about 284.5 g of the about 21 wt. % water soluble resin of Attachment F with 115.5 g of water. The calculation for preparing the 15% binder is shown (and highlighted) on the right side of the first page of Attachment G. The second page of Attachment G presents the "dry" and "hot/wet" tensile test results for "Binder 5." The raw data for the dry tensile testing of 12 replicates and the hot/wet tensile testing of 12 replicates are both setoff with yellow highlighting and the data respectively correspond to average tensile strengths of 41.7 pounds and 33.3 pounds (shown below the horizontal line on Attachment G). Based on these results we concluded that the water soluble resin had been successfully used and thus was useful as a binder for glass fibers.

14.) THAT Attachment H are the laboratory notebook records (notebook pages 228G67 (two sides) and 228G68) of two separate free radical polymerizations of maleic anhydride (MA) and hydroxyethyl acrylate (HEA) in the presence of allyloxy (allyloxy) propane diol (APD) as the chain transfer agent. As shown on pages 1 and 2 of Attachment H, about 0.92 mole of maleic anhydride was added into a reaction vessel containing water. The contents of the reactor were heated to 70 °C and held at that temperature for 30 minutes. About 15 parts (about 0.11 mole) of the chain transfer agent (APD) then was added. One (1) part of the free radical catalyst azodiisobutyronitrile (AIBN) was added and then over a one hour period, about 0.69 mole of hydroxyethyl acrylate was added into the reactor and polymerization ensued. Once the

HEA was added, the reaction was allowed to remain at the elevated temperature for approximately an additional 1½ hours to yield a resin product having a solids content of about 28 wt. %. The relative mole ratio of the two monomers (MA:HEA) was 1.33:1.0 to provide a mole ratio of –COOH:-OH of about 2.7:1.0. In the second synthesis, recorded on page 3 of Attachment H, about 2.75 moles of maleic anhydride was added into a reaction vessel containing water. The contents of the reactor were heated to 72 °C and held at that temperature for 30 minutes. About 45 parts (about 0.34 mole) of the chain transfer agent allyloxy-1,2-propanediol (allyloxy propane diol or APD), then were added. 2.5 parts of the free radical catalyst azodiisobutyronitrile (AIBN) were added and then over a 1½ hour period, about 2.68 moles of hydroxyethyl acrylate were added into the reactor. Once the HEA was added, the reaction was allowed to remain at the elevated temperature for approximately an additional ½ hour to yield a water soluble resin having a solids content of about 32 wt. %. The relative mole ratio of the two monomers (MA:HEA) was 1.03:1.0 to provide a mole ratio of –COOH:-OH of about 2.1:1.0.

15.) THAT Attachment I (notebook records 228G69 and 228G70) documents the preparation of several binder formulations for preparing handsheets and the results of conducting the testing referred to in paragraphs 9.) and 10.) on the handsheets. For purpose of the present invention, the binder labeled 3 ("Binder 3") was prepared using the first resin prepared in accordance with paragraph 14.); while the binder labeled 4 was prepared using the second resin prepared in accordance with paragraph 14.). 400 grams of binder containing 20 wt. % resin solids was prepared from both resins by mixing, in the first case, about 290 g of the about 28 wt. % resin of Attachment H with 110 g of water and in the second case by mixing about 256 g of resin with 144 g water. The calculations for preparing the 20% by weight solids binders are shown (and highlighted) on the first page of Attachment I. The second page of Attachment I

presents the "dry" and "hot/wet" tensile test results for "Binder 3" and "Binder 4," as well as for Binder 2, a phenolic control. The raw data for the dry and hot/wet tensile testing of 12 replicates are both setoff with yellow highlighting and the data respectively correspond, for Binder 3 to average tensile strengths of 36.9 pounds and 28.6 pounds and for Binder 4 to average dry and hot/wet tensile strengths of 39.7 pounds and 29.2 pounds. The PF resin binder control exhibited dry and hot/wet tensile strengths of about 38.4 and 31.5 pounds respectively. Based on these results, we concluded that the water soluble resins of Attachment H had been successfully used and thus were useful as binders for glass fibers.

16.) THAT the dates recorded on each of the documents in Attachments A-I have been removed.

We hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: December 20, 2005 By: _____
Ramji Srinivasan

Date: 12/20/05 By: _____
Cornel Hagiopol

Date: 12/22/05

By: Natasha R. Bailey

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LE Copolymer from MA/HEA + Allyloxy propanediol Book No. 28668

m Page No.

aw Material	Weight (grams)	Moles
water	140.0	
llyloxy-1,2-propanediol	45	0.3404
laleic Anhydride	270	2.753
ydroxyethyl Acrylate	313	2.066
AIBN	2.5	

Time	%NV	Notes
9:38		Charged water
9:47		added MA + warmed to 72C (introduced nitrogen) data here
10:14		held at 72C for 30 min; prepared HEA+H2O in funnel
10:44		Charged Allyloxypropane diol
10:48	20.01	sampled for solids
11:03		added AIBN
11:10		Added HEA + H2O Over at least 90 min
1:20	30.36	Sampled for solids; allowed cook to stir for 30 more minutes
1:50	31.47	Sampled for solids
2:12		Cooled allowed polymer to slowly cool
3:18	31.89	sampled for solids

RI
pH
NV
Viscosity

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Witnessed & Understood by me.	Date	Invented by	Date
			redacted
		Recorded by Natasha Bird	

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Scaling up of 226G67. 226G.

To repeat 226G67 (Natasha) but increasing the quantity for laboratory testing.

Material.	wt.		
MA.	98.06	270.00	2.7534
APN :	132.96	45.00	0.3407
HEA :	116.12	240.00	2.0668.
water :		1500.00	
AIBN :		3.00	

9.30 T: 22°C Maleic anhydride / HEA Charged
9.50 T: 23°C Allyl 1,1-Propanediol charged
10.00 T: 23°C.
10.05 weighed out HEA
10.15 22°C. AIBN Charged
10.17 22°C HEA Charge started
10.30 22°C
10.45 23°C. 1/2 HEA Charged
10.50 24°C.
11.05 HEA addn completed
NV: 29.26 g.
11.30 NV: 29.48
11.55 NV: 29.44 g. Run stopped

redacted redacted

646

GP RESINS
redacted 10:
LHCO
SAMPLE 199

Dry Time: 01
Max Temp: 1(
Initial: 1.

27.54

Prepartion of cationic version of MA/HEA copolymer
Copolymerization of maleic anhydride with Hydroxyethyl acrylate
Using THIO GLYCOL as chain transfer agent

Raw Material	Weight	Moles	wt %
Water	400.00		71.8623
Maleic Anhydride	60.00	0.61	10.7600
Hydroxyethyl acrylate	73.15	0.63	13.1183
Mercapto Ethanol	1.00		0.1793
MTA	20.77	0.10	3.7248
AIBN	0.70		0.1255
Mercapto Ethanol	1.00		0.1793
	556.62		99.95

[2(Methacrolooxy)ethyl]trimethylammoniumChloride

Charge water
Charge Maleic anhydride
Warm to 72-73C
Hold for 30 minutes at this temperature
In the meanwhile Weigh out HEA/MTA in a addition funnel. Add 1.0 g of MERCAPTOETHANOL to it and mix
Check Slolds (~15%)
Add AIBN
Add Mercaptoethanol 1.0 g to the reaction mixture
Immediately Start adding Hydroxyethyl acrylate slowly in drops. (atleast 60 minutes)
Maintain temperature at 72- 73 C

Check solids every 30 minutes until no increase in solid is seen .
Target solids ~25%

TITLE Ran of MA/HEA with cationic monomer Book No. 236646

From Page No.

To Make a cationic version of MA/HEA copolymer
Using ~~MAA~~ methacryloxy ethyl trimethyl ammonium chloride.
Using ~~beta~~ mercapto ethanol as CT agent.

8.30		water charged
8.35		MA added.
8.40	23°C	Heat started to 72°C.
8.55	55°C.	
9.00	70°C.	Solids : 14.87. (Expted: ≈ 15%)
		HEA/~~MAA~~MTA weighed int addn funnel.
		Add 1g beta mercapto ethanol.
9.15		Mercapt ethanol (1.0 g added to kettle.
9.20		AIBN added
9.24	72°C	Addn of Monomer started
9.35	74°C.	
9.40	74°C	1/2 the monomer added
10.00	73°C.	
10.15	76°C.	Add monomer added.
10.20	73°C	NV: 27.54
10.40	72°C	NV: 29.54 / 29.53.
		Ran stopped by cooling down

Product came well & does not swell in water.

NV: 29.54
pH: 1.45
V: 15.5 → redacted
pH: 0.90

To Page No.

Witnessed & Understood by me,	Date	Invented by	Date
P C Bryan	redacted	Recorded by	redacted

– GWS

Prepartion of cationic version of MA/HEA copolymer
Copolymerization of maleic anhydride with Hydroxyethyl acrylate.
Using THIO GLYCOL as chain transfer agent

Raw Material	Weight	Moles	wt %
Water	400.00		71.7334
Maleic Anhydride	60.00	0.61	10.7600
Hydroxyethyl acrylate	73.15	0.63	13.1183
Mercapto Ethanol	2.00		0.3587
MTA	20.77	0.10	3.7248
AIBN	0.70		0.1255
Mercapto Ethanol	1.00		0.1793
	557.62		100.00

[2(Methacrolooxy)ethyl]trimethylammoniumChloride

Charge water
Charge Maleic anhydride
Warm to 72-73C
Hold for 30 minutes at this temperature
In the meanwhile Weigh out HEA/MTA in a addition funnel. Add 2.0 g of MERCAPTOETHANOL to it and mix
Check Sloids (~15%)
Add AIBN
Add Mercaptoethanol 2.0 g to the reaction mixture
Immediately Start adding Hydroxyethyl acrylate slowly in drops. (atleast 60 minutes)
Maintain temperature at 72- 73 C

Check solids every 30 minutes until no increase in solid is seen.
Target solids ~~25%~~ 30%.

TITLE To make a Cationic version of 228G72 Project No.____ Book No. 136445

From Page No.__

To make a co-polymer using MA/HEA/MTA

This will be a repeat of 228G72 (Natalie's Note book)
The difference is that a cationic monomer methoxyethyl-trimethyl ammonium chloride is added at 0.1M to give a cationic charge to the polymer.

11.20	22°C	water added
11.25	20°C	MA added
11.26		Heat started / N2 started.
11.36	40°C	
11.45	72°C.	HEA/MTA / Mercapto ethanol weighed on ~~added~~.
11.50	70°C.	Mercapto ethanol added to kettle.
11.56	72°C	AIBN added
12.00	72°C	Addn of monomer started.
12.30	72°C	1/2 " " done
1.00	73°C	Addn. completed
1.15	75°C.	NV- 28.41
2.00		NV- 29.54
2.10		NV- 29.55.
		Rxn stopped.

The viscosity is v. low.

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Witnessed & Understood by me,	Date	Invented by	Date
[signature]	redacted	[signature] Recorded by	redacted

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Raw Material	Weight	Moles
Water	400	
Maleic Anhydride	50	0.52
Hydroxyethylacrylate	116.12	0.78
COPS	20.05	0.05
AIBN	0.7	

Time	
10:35	Charged water tran Nitrogen to get rid of free radicals for 20 min.
10:55	Charged MA + warmed to 72C
11:17	hold for 20 min
11:37	added COPS
11:45	Added AIBA + Hydroxyethyl acrylate (over 1hr)
12:45	let stir 30 min.
1:30	raised temp to 78C
	added 218 g H2O

% NV = 15.03%

The rxn did not go to Completion The viscosity was too high & the rxn was stopped. Added water

wt of Q container = 34.83

wt of Q container + polyester = 799.21

764.17g

Copolymerization of maleic anhydride with Hydroxyethyl acrylate.
(Using Sipomes COPS as Chain Transfer agent)

Raw Material	Weight	Moles
Water	400	
Maleic Anhydride	50	0.52
Hydroxyethyl acrylate	116.12	0.78
COPS	10.05	0.05
AIBN	0.7	

20g ←

wt of kettle + polyester = 1127.11

wt of kettle = 362.44

764.67

redacted

Charge water
Charge Maleic anhydride
Warm to 72-73C
Hold for 20 minutes at this temperature
Add COPS
Add AIBN
Immediately Start adding Hydroxyethyl acrylate slowly in drops. (atleas 60 minut → for 1 hr
Maintain temperature at 72- 73 C
After addition is complete let stir for 30 more minutes.

Witnessed & Understood by me,	Date	Invented by	Date
	redacted	Recorded by	redacted

Handsheet Study

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~~500g (binder) x 20% solids = 100g solid~~

~~228649 @ 29.52%: $\frac{100}{.2952}$ = 338.75g solid~~

~~500 - 338.75 = 161.25 g water~~

500 x 15% solid = 75g solid

$\frac{75}{.20}$ = 375g solid

500 - 375 = 125g water

500g binder x 20% solids = 100g solids

• 228644 @ 49.02%: $\frac{100}{.4902}$ = 204.00g solid

500 - 204.00g = 296g water

500g x 15% solids = 75g solid

$\frac{75}{.20}$ = 375g solid

500 - 375 = 125 g water

500g binder x ~~20%~~ 15% solids = ~~100g~~ 75 solids

• 228650 @ 15.03: $\frac{75g}{.1503}$ = 499.00g solid

500 - 499.00 = 1g water

500g x 15% solid = 75g

kirolynol $\frac{75}{~~.40.75~~ .30}$ = ~~[illegible]~~ 250 solid

500 - 250 = ~~375.95~~ 250 g water

236636 @ 5%

(5) 228650 @ 5%

500g x .05 = 25

$\frac{25}{.1503}$ = 166.33g solid

500 - 166.33 = 333.67g water

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Witnessed & Understood by me: [signature] Date: redacted

Invented by / Recorded by: [signature] Date: redacted

Weight of binders @220 for 2 min

15% Binder 1 smokes	15% Binder 2 smokes	15% Binder 3 smokes (4 minutes)	Binder 4	5% smokes Binder 5 (2 min)
8.50	7.63	11.6	7.59	8.63
8.49	7.97	10.02	7.74	7.78
8.40	7.79	10.20	8.15	7.99
8.42	8.24	12.32	8.27	7.78

Tensile Strength

dry

Binder 1	Binder 2	Binder 3	Binder 4	Binder 5
60.3	33.2	66.9	12.8	29.5
57.3	32.6	53.9	9.5	19.2
63.6	38.3	94.5	12.7	25.6
40.7	21.1	88.1	16.6	19.1
45.1	23.3	95.9	10.8	24.9
37.1	23.1	49.3	16.8	28.5
48.7	28.4	85.1	15.2	39.1
37.5	34.5	61.7	17.8	29.9
40.8	20.8	57.8	13.1	29.5
20.2	26.4	61.6	18.4	29.9
57.1	26.2	55.2	16.0	29.5
62.0	35.1	72.9	12.6	12.4
57.7	22.8	61.6	16.7	19.2
52.0	40.6	53.3	11.6	30.2

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Witnessed & Understood by me.	Date	Invented by	Date
[redacted]	redacted	Recorded by Natasha Buck	redacted

228653

Tensile Strength (cont.)

wet nder 1	Binder 2 (236438 / 228444)	Binder 3	Binder 4 (MA/HEA 236436 @ 5l.)	Binder 5 (228450 MA/HEA)
1.2	3.6	32.1	8.9	7.2
0.4	2.8	34.4	6.4	5.9
0.2	3.0	29.8	7.0	8.1
6.1	5.1	27.5	9.8	5.2
3.6	4.7	23.9	9.6	5.0
4.4	4.2	37.7	8.9	4.2
2.6	6.1	26.8	12.4	5.5
2.1	3.0	32.5	7.4	1.1
7.9	3.4	42.3	6.3	1.3
6.9	3.6	49.7	6.2	1.7
.3	5.6	31.9	7.3	1.3
.9	6.2	40.7	7.7	2.1
1.4	6.2	40.9	6.8	1.1
.2	5.7	35.3	9.6	1.9

sed & Understood by me

Date redacted

Witnessed by

Date redacted

Title: Copolymer of Itaconic Acid / ~~MAA~~ HEA + Alloxypropanediol Project No. Book No. 228G64

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Raw Material	Weight	Moles
Water	400	
Alloxy 1,2 propane diol	15.9	0.13877
Itaconic Acid	58.3	0.5945
Hydroxyethyl Acrylate	92.8	0.7972
AIBN	0.7	

Time	%NV	Notes
7:15		Charged water
7:18		Charged Allyloxypropane diol ; mixture clear
7:30		Charged Itaconic Acid
7:42		raised temp to 38C to dissolve Itaconic Acid
8:25	14.80	warmed to 72C
8:43		added AIBN + started HEA over 60min
9:50	18.25	HEA addition complete + sample taken
10:30	17.27	
11:00	18.56	
11:30	18.12	
12:00	20.13	
12:30	20.87	
1:10	22.21	
1:30	22.26	
1:54		raised temp to 76C
2:00	23.22	
2:16		added .214g AIBN
2:35	22.00	
3:00	21.40	

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Witnessed / Understood by me	Date	Witnessed by	Date
redacted	redacted	Approved by	

redacted

Co-polymer of Itaconic acid Maleic anhydride and Alloxy propanediol

Raw Material	Weight	Moles
Water	400.0	-
Alloxy1,2 propane diol	15.9	0.13877
Itaconic acid	58.3	0.5945
Hydroxyethyl acrylate	92.8	0.7972
AIBN	0.7	

7:15 Charge water

7:18 Charge Allyloxypropane diol

Stir for few minutes. Make sure the reaction mixture is clear.

7:30 Add Itaconic acid redacted

After all Itaconic acid has dissolved. Check Solids.

Weigh out HEA into Addition funnel, Add 30 g of water to it and keep it ready

8:25 Warm to 72 C

9:43 Add AIBN

Immediately Start adding Hydroxyethyl acrylate slowly in drops. (atleast 60 minutes) redacted

Maintain temperature at 72- 73 C

After addition is complete let stir for 30 more minutes.

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Binder 2 228G60

228G60 @ 16.30% solid

400 x .15 = 60g

$\frac{60g}{.1630}$ = 368.10 g solid needed

400 - 368.10 = 31.9g H_2O needed

Binder 1 QRXP

~~228G6~~ QRXP @ 40.91% solid

400 x .15 = 60g

$\frac{60}{.4091}$ = 146.66g solid needed

400 - 146.66 = 253.34g H_2O needed

Binder 3 228G61

228G61 @ 20.15% solid

400 x .15 = 60g

$\frac{60}{.2015}$ = 297.77g solid needed

400 - 297.77 = 102.23g H_2O needed

Binder 4 228G62

228G62 @ 19.80% solid

400 x .15 = 60g

$\frac{60}{980}$ = 303.03g solid needed

400 - 303.03 = 96.97g H_2O needed

Binder 5 228G64

228G64 @ 20.87% solid

400 x .15 = 60g

$\frac{60}{.2087}$ = 284.49g solid

400 - 284.49g = 115.51g H_2O needed

Binder 6 Interpolymer

Interpolymer @

Read & Understood by me.

Date redacted

Invented by

Witnessed by

Date redacted

TLE Handsheet Study Book No 228-66

Dry hot/wet

Binder 1		Binder 2		Binder 3	
54.0	3.3	41.1	37.3	27.7	35.5
61.2	3.1	36.7	29.4	short strip ~~11.0~~	34.4
37.9	4.2	27.4	17.41	29.4	37.6
73.9	4.3	38.2	27.0	17.9	33.0
62.3	4.2	31.9	23.1	47.7	37.0
50.3	2.7	23.6	25.1	28.9	21.3
34.8	1.8	19.4	21.7	33.6	28.3
77.6	4.5	16.3	141.6	25.2	35.9
79.8	3.6	41.6	32.9	17.8 ~~12.8~~	27.5
46.1	8.2	32.1	27.9	17.0	32.4
49.5	6.4	12.5	24.8	19.0	37.3
30.1	1.8	14.3	16.4	31.4	12.5 short strip
55.63	4.01	27.93	24.80	25.13	31.02

Binder 4		Binder 5		Binder ~~5~~ 6 Interpolymer	
58.7	31.5	46.7	28.1	78.6	12.3
22.8	33.1	50.4	38.1	43.2	8.1
56.6	49.0	35.9	35.7	46.9	10.1
40.7	28.7	42.5	28.5	49.0	3.7
40.5	58.1	57.1	50.6	47.6	10.1
60.7	40.7	26.4	32.8	47.1	8.2
25.2	36.9	48.5	25.8	55.4	11.2
67.3	31.5	35.7	38.9	60.6	5.6
43.6	27.1	48.6	47.4	68.5	7.8
62.2	25.0 short strip	short strip ~~19.1~~	29.3	54.7	8.0
21.3	30.5	45.1	32.0	34.4	4.3
58.1	42.6	39.6	12.3 short strip	53.2	7.5
46.48	36.23	41.72	33.29	53.27	8.08

To Page No.....

Witnessed & Understood by me.	Date	Invented by	Date
	redacted	Recorded by	redacted

OIPE IAP40 DEC 28 2005 PATENT & TRADEMARK OFFICE

Project No.

TITLE: Copolymerization of MA/HEA and APD Book No. 228967

From Page No.

Raw Material	Weight	Moles	
water	500		
Allexy propane diol	15	0.1136	
Maleic Anhydride	90	~~0.1978~~	0.9178
Hydroxyethyl Acrylate	80	0.688	
AIBN	1		

Time	Notes	Solid %
10:45	Charged water	
10:48	Added MA & warmed to 70 C	
11:07	Held at 70C for 30 min	
11:37	pH = 1.00	
11:42	Charged APD ~~pH=1.03~~	
11:45	pH = 1.09 ~~added AIBN~~	
11:48	added AIBN + added HEA over 1hr (maintained temp 72-73C)	
12:48		26.77
1:08		27.84
1:28		28.04
1:48		27.87
2:08		28.16
2:28		27.95

2:08

GP RESINS
13:28
LHC
SAMPLE 97
redacted
Dry Time: 02:20
Max Temp: 105 C
Initial: 2.2451g

28.16% S

2:

GP RESINS
redacted 13:
LHC
SAMPLE 98
Dry Time: 02:3
Max Temp: 105 C
Initial: 2.3023

27.95% S

wt of sample container = 13.31g
wt of sample = 19.87g

RI = 1.3729
Viscosity = 24
pH = ~~7~~ NB 1.63

Witnessed / Understood by	Date	Invented by	Date
	redacted	Recorded by	redacted

12:98

redacted
Copolymerization of MA/HEA and APD

Raw Material	Weight	Moles
Water	500 -	
Alloxy propane diol	15	0.1136
Maleic anhydride	90	0.1978
Hydroxyethyl acrylate	80	0.688
AIBN	1	

GP RESINS
redacted 12:10
LHC
SAMPLE 91

Dry Time: 02:41
Max Temp: 105 C
Initial: 2.3608g

26.77% S

0:45 Charge water
10:48 Add Maleic anhydride
Warm to 70 C Hold fro 30 minutes, Check pH
Weigh out HEA into Addition funnel, Add 30 g of water to it and keep it ready
Charge APD Check pH
Add AIBN
Immediately Start adding Hydroxyethyl acrylate slowly in drops. (atleas 60 minutes)
Maintain temperature at 72- 73 C,
After addition is complete Check solids every 20 minutes Until target solids is reached

1.08

GP RESINS
redacted 12:28
LHC
SAMPLE 92

Dry Time: 02:20
Max Temp: 105 C
Initial: 1.5351g

27.84% S

1:28

GP RESINS
redacted 12:54
LHC
SAMPLE 95

Dry Time: 02:08
Max Temp: 105 C
Initial: 1.9670g

28.04% S

1:48

GP RESINS
redacted 13:08
LHC
SAMPLE 96

Dry Time: 02:32
Max Temp: 105 C
Initial: 2.6204g

redacted 27.87% S

Bal

redacted

Natasha

From Page No.

Raw Material	Weight (grams)	Moles
Water	1400	
Allyloxy-1,2-propanediol	45	0.3404
Maleic Anhydride	270	2.753
Hydroxyethyl Acrylate	313	~~2.866~~ 2.6755
AIBN	2.5	

Time	%NV	Notes
9:38		Charged water
9:47		added MA + warmed to 72C (introduced nitrogen start heat)
10:14		held at 72C for 30 min; prepared HEA+H_2O in funnel
10:44		charged Allyloxypropane diol
10:48	20.01	sampled for solids
11:03		added AIBN
11:10		Added HEA + H_2O Over at least 90 min
1:20	30.36	Sampled for solids; allowed cook to stir for 30 more minutes
1:50	31.47	Sampled for solids
2:12		~~Cooled~~ allowed polymer to slowly cool
3:18	31.89	Sampled for solids

RI = 1.3791
Viscosity: 75
pH = ~~8.08~~ 1.63

To Page No.

Witnessed & Understood by me.	Date	Invented by	Date
	redacted	Recorded by Natasha Baid	redacted

Title Handsheet Study Book No. 228669

OIPE IAP40
DEC 2 8 2005
PATENT & TRADEMARK OFFICE

From Page No.___

~~Binder 3~~ Phenolic Cure...

~~Binder 3 228667~~

* Binder 1 = QRXP @ 40.91% solid

400g x .20% = 80

$\frac{80}{.4091}$ = 195.55g solid

400 - 195.55 = 204.45g H_2O

* Binder 2 = Phenolic control @ 32.25% solid

400g x .20% = 80g

$\frac{80}{.3225}$ = 248.06g solid

400 - 248.06g = 151.94g H_2O

* Binder 3 = 228667 @ 27.63% solid

400g x .20% = 80

$\frac{80}{.2763}$ = 289.54g solid

400 - 289.54 = 110.46g H_2O

* Binder 4 = 228668 @ 31.20% solid

400g x .20% = 80

$\frac{80}{.3120}$ = 256.41g solid

400 - 256.41g = 143.59g H_2O

To Page No.___

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TITLE Handsheet Study

Project No.

Book No. 228670

From Page No.

Binder 1 QRXP

	Hot/wet
19.4	7.8
47.8	3.5
56.7	6.2
61.3	9.5
18.9	7.3
67.7	5.2
45.1	5.1
49.8	8.4
22.8	9.2
47.7	4.3
68.6	4.2
59.8	5.8

Binder 2 Phenolic Control

	Hot/wet
18.8	41.8
44.5	39.5
45.5	31.3
54.7	22.0
11.4	40.8
33.2	35.3
41.0	19.5
33.9	29.1
14.5	35.1
52.9	37.3
49.3	27.0
60.6	19.8

Binder 3 228667 heavy smoke

	Hot/wet
16.0	38.9
46.1	26.9
40.4	29.3
39.5	30.6
14.6	22.6
37.2	23.3
35.1	26.0
52.4	29.7
17.7	40.6
52.3	28.1
46.4	22.6
44.7	24.7

Binder 4 228668 heavy smoke

	Hot/wet
14.9	38.0
52.2	40.7
40.4	19.2
49.6	21.6
17.7	34.6
43.6	34.1
54.7	31.1
57.9	23.2
15.1	32.9
47.1	25.5
39.2	26.1
39.2	31.4

To Page No.

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